

Spontaneous Stratification in Granular Mixtures

Hernán A. Makse,¹ Shlomo Havlin,^{1,2} Peter R. King,³ and H. Eugene Stanley¹

¹*Center for Polymer Studies and Physics Dept., Boston University, Boston, MA 02215 USA*

²*Minerva Center and Department of Physics, Bar-Ilan University, Ramat Gan, Israel*

³*BP Exploration Operating Company Ltd., Sunbury-on-Thames, Middx., TW16 7LN, UK*
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Granular materials [1–5] size segregate when exposed to external periodic perturbations such as vibrations [6–12]. Moreover, mixtures of grains of different sizes spontaneously segregate in the absence of external perturbations: when a mixture is simply poured onto a pile, the large grains are more likely to be found near the base, while the small grains are more likely to be near the top [13–20]. Here, we report a novel spontaneous phenomenon arising when we pour a mixture between two vertical plates: the mixture spontaneously stratifies into alternating layers of small and large grains whenever the large grains have larger angle of repose than the small grains. In contrast, we find only spontaneous segregation when the large grains have smaller angle of repose than the small grains. The stratification is related to the occurrence of avalanches; during each avalanche the grains comprising the avalanche spontaneously stratify into a pair of layers, with the small grains forming a sublayer underneath the layer of large grains.

Our experimental system consists of a vertical “quasi-two-dimensional” cell with a gap of 5 mm separating two transparent plates (made of plexiglass, or of glass) measuring 300 mm × 200 mm (see Fig. 1a). We choose this quasi-two-dimensional geometry since, by using this setup, the internal features of the avalanche process can be easily visualized, both statically and dynamically. To avoid the effects of electrostatic interaction with the wall, the wall is cleaned with an antistatic cleaner.

In a first series of experiments, we close the left edge of the cell leaving the right edge free, and we pour, near the left edge, an equal-volume mixture of white glass beads (mean size 0.27 mm, spherical shape, and repose angle 26°), and red sugar crystals (typical size 0.8 mm, cubic shape, and repose angle 39°). Figure 1a shows the result of the first series of experiments. We note two features:

- (i) *Spontaneous Stratification.* We see the formation of alternating layers consisting of small and large grains—with a “wavelength” of about 1.2 cm.
- (ii) *Spontaneous Segregation.* We find that the smaller grains segregate near the left edge and the larger grains segregate furthest from it and near the base [13–20].

In a second series of experiments, we confirmed the results of these initial experiments by testing for stratification and segregation using a mixture of grains of same density, consisting of fine sand (typical size 0.4 mm) and coarse sand (typical size 1 mm), suggesting that the density of the grains may not play an important role in stratification.

In all the above experiments we used mixtures composed of two types of grain with different shape, and therefore with different angles of repose. In particular we obtain stratification (plus segregation) when we use larger cubic grains and smaller spherical grains: the angle of repose of the large species is then larger than the angle

of repose of the small species. Otherwise we obtain only segregation and not stratification when the large grains are less faceted than the small grains, i.e., the large grains have smaller angle of repose than the small grains.

To confirm this, we performed a series of experiments using mixtures of irregular shaped sand grains (repose angle 35°, and mean size 0.3 mm), and spherical glass beads (repose angle 26° *smaller* than the repose angle of the sand grains). We find that stratification (plus segregation) occurs for two different experiments using spherical beads of size 0.07 mm and 0.11 mm (so that the larger grains have larger repose angle). In contrast, we obtain only segregation *but not stratification* for two experiments using spherical beads of size 0.55 mm and 0.77 mm (so that the larger grains have smaller repose angle). In all cases the segregation of grains occurs with the smaller grains being found near the left edge of the cell and the larger grains near the base of the cell. These results suggest that the phenomenon of segregation is always expected when pouring a granular mixture of grains of different sizes, no matter the values of the angles of repose of the species. However, the phenomenon of stratification is only expected when the large species have larger angle of repose than the small species.

Additionally, we performed a series of experiments in which we find similar stratification by using different mixtures of differing size ratio between large and small grains (1.66, 2.1, 2.25, 3.25, and 6.66), suggesting that the phenomenon occurs for a broad regime of grain size ratios. We find a similar stratification when we double the gap between the vertical plates of the cell and simultaneously double the flow rate of grains.

We propose a physical mechanism responsible for the observed stratification that is related to the fact that not one but rather a pair of layers is formed in the course of each avalanche. When the flow of grains reaches the base of the pile, we find that the grains develop a profile characterized by a well-defined “kink”, at which the grains

are stopped (see Fig. 1b); but we find that the small grains stop first, so a pair of layers forms with the small grains *underneath* the large grains. As more grains are added, the kink appears to move upward in the direction opposite to the flow of grains. Once the kink reaches the top, the pair of layers is complete and the cycle is then repeated: a new avalanche occurs, the kink develops, and a new pair of layers forms.

The “wavelength” of a pair of layers λ can be determined by the mean value of the downward velocity v of the rolling grains during an avalanche, the upward velocity v' of the kink, and the thickness of the layer of rolling grains R_0 during the avalanche. If the volume of grains in an avalanche scales approximately as the volume of grains in a well-formed kink, we predict $\lambda \simeq R_0(v + v')/v'$, and we confirm this relation experimentally.

To test this physical mechanism by computer simulation we consider a mixture comprising small grains of width one pixel and of height H_1 , and large grains, also of width one pixel but of height $H_2 > H_1$. To generate an equal-volume mixture, we randomly drop a small grain with probability $p \equiv H_2/(H_1 + H_2)$, and drop a large grain with probability $1 - p$ (see Fig. 2).

In critical phenomena, it is often useful to first develop a “mean field” type model (in which, for example, spin orientation is determined by a macroscopic variable, the net magnetization), before devising models in which spin orientation is determined by the microscopic quantities such as the orientations of the other spins comprising the system. In this spirit, we focus first not on the “microscopic” grain motions, but rather on the “macroscopic” angle of the sandpile, whose value alternates in time between the maximum angle of stability θ_m which defines the onset of an avalanche, and the angle of repose θ_r which defines the end of the avalanche [21,22]. Using this model (described in the legend of Fig. 2), we find a morphology that displays both segregation and stratification.

In addition to the simplest “mean field” approach, we develop a model [23] in which we treat the individual grain motion in accord with microscopic rules that depend not on the macroscopic angle of the sandpile, but rather on the local angles formed between each grain and its neighbors. Specifically, the dynamics of the small and large rolling grains are governed by the critical angles of repose corresponding to the interactions between the rolling grain and the static grains of the sandpile surface. This model incorporates the experimental fact that grains segregate because large grains roll down more easily on top of small grains than small grains on top of large grains (for rolling *large* grains on top of a surface of small grains the surface appears smoother than for rolling *small* grains rolling on top of a surface of large grains). Thus the model correctly predicts that the small grains form a sublayer beneath the large grains. We find stratification, as in the simplest “mean field” model, and also find that the profile of the sandpile displays a kink at which rolling grains are stopped—just as in the experiment.

Next we test the above principles by generalizing from two grain sizes to three. The experiment results in stratification with three layers, with the finest grains on the bottommost of each triplet of layers and the coarsest grains on the topmost layer (see Fig. 1c where the same experimental setup of Fig. 1a is used to obtain an alternation of *three* layers of grains of *three* different sizes: 0.15 mm, 0.4 mm, and 0.8 mm). Experiments using a continuum size distribution are ongoing, since geological rock formations (which also display stratification) generally occur in the presence of a continuum distribution of grain size.

As another test of the proposed physical mechanism, we note that the case of spherical grains should not lead to stratification because the angles of repose of the large and small grains are the same. We confirm this prediction experimentally. The case of spherical grains was also studied by Williams [24–26]; his results (showing segregation plus a hint of stratification) differ from our results (showing only segregation), presumably because his grains were not quite spherical—i.e., the repose angle of the large and small “spheres” were slightly unequal.

Finally, we note that Bouteux and de Gennes have recently made considerable progress [27] in developing a general theoretical framework [28,29] to treat the case of granular flows of two different grains. Their conclusions [27–29] are consistent with the experiments presented here.

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FIG. 1. Experimental results: **a**, Typical result of the first series of experiments, showing the formation of successive layers of fine and coarse grains (here the white grains are glass beads of average diameter 0.27 mm, while the larger grains are sugar crystals of typical size 0.8 mm). We clean the walls of the cell with antistatic cleaner, in order to avoid the effects of electrostatic interaction between the grains and the walls. We pour the equal-volume mixture near the left edge between two transparent vertical plates separated by a gap of 5 mm. We obtain stratification with a wavelength $\lambda \simeq 1.2$ cm. **b**, Close-up photograph of the kink where the grains stop during an avalanche. The small white grains stop first, and then the large red grains; hence the small grains form a sublayer underneath the large grains. **c**, Stratification obtained using a mixture of three different types of grains: nearly spherical glass beads (0.15 mm, angle of repose 26°), blue sand (0.4 mm, angle of repose 35°), and red sugar crystals (0.8 mm, angle of repose 39°). We notice the grading (from bottom to top) in a triplet of layers: small (white), medium (blue), and large (red) grains. **d**, Close-up of **c**, field of view 40 mm \times 40 mm.

FIG. 2. Results of modeling: **a**, The dynamics of the simplest “mean field” model are illustrated by this example with two sizes $H_1 = 1$ (white) and $H_2 = 2$ (red), and threshold slopes $s_r \equiv \tan \theta_r = 2$ and $s_m \equiv \tan \theta_m = 3$. Suppose that, at a given instant, the sandpile is at the critical slope for repose s_r . To define the dynamical rules for the arriving grains, we consider the slope $s_i \equiv h_i - h_{i+1}$, where h_i denotes the height of the sandpile at coordinate i . We deposit a grain near the first column at the left edge of the lattice, where the actual column position is chosen from a narrow Gaussian probability distribution centered at the wall edge. The non-zero width of this Gaussian mirrors the fact that grains often bounce after reaching the pile. Newly arriving grains accumulate on the sandpile profile, following dynamics governed by the critical slope s_m ; thus a grain moves from the initial landing point at column i to column $i + 1$ if the slope $h_i - h_{i+1}$ is larger than s_m , then moves from column $i + 1$ to column $i + 2$ if $h_{i+1} - h_{i+2} > s_m$, and so forth. The grain stops at the first column k with $h_k - h_{k+1} \leq s_m$. Another grain is now added, and the same rules are followed. **b**, This entire process continues until a grain reaches the substrate at the furthest right column of the pile for first time (grain 8 in this figure). Now, since $s_i > s_m$ for all columns i , the sandpile has become “unstable”. We note that s_i is calculated considering also the size of the rolling grain; as a result, the large grains more readily reach a slope that exceeds the two critical slopes s_r and s_m . **c**, We allow the sandpile to relax toward the repose slope s_r by moving each of the grains with slope larger than s_r to the nearest column satisfying $h_i - h_{i+1} \leq s_r$. Now the deposition starts again, and we iterate the algorithm until a large sandpile (of typically 10^5 grains) is formed. We can obtain stratification with constant “wavelength” by stopping the accumulation process when a grain reaches for first time the column $l^{1/2}$, where l denotes the furthest right column of the pile. **d**, Image obtained with the simplest “mean field” model (for parameters $H_1 = 1$, $H_2 = 2$, $s_r = 4$, and $s_m = 5$); the smaller grains are white and the larger grains red. We find stratification and also reproduce the “kink” mechanism explained in the text when we improve upon the simplest “mean field” model by including four different angles of repose to take into account the fact that the angle of repose depends on the concentration of grains at the surface of the pile [23].

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